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LLNL NESHAPs 2012 Annual Report

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**U.S. Department of Energy
Radionuclide Air Emission Report for 2012
(in compliance with 40 CFR 61, Subpart H)**

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Executive Summary

Lawrence Livermore National Security, LLC operates facilities at Lawrence Livermore National Laboratory (LLNL) where radionuclides are handled and stored. These facilities are subject to the U.S. Environmental Protection Agency (EPA) National Emission Standards for Hazardous Air Pollutants (NESHAPs) in Code of Federal Regulations (CFR) Title 40, Part 61, Subpart H, which regulates radionuclide emissions to air from Department of Energy (DOE) facilities. Specifically, NESHAPs limits the emission of radionuclides to the ambient air to levels resulting in an annual effective dose equivalent of 10 mrem (100 μ Sv) to any member of the public. Using measured and calculated emissions, and building-specific and common parameters, LLNL personnel applied the EPA-approved computer code, CAP88-PC, Version 1.0, to calculate the dose to the maximally exposed individual member of the public for the Livermore site and Site 300.

In 2012, LLNL maintained its compliance with 40 CFR 61, Subpart H. All radioactive air emissions resulted in calculated doses well below the annual 10 mrem (100 μ Sv) site-wide standard. The annual doses to the site-wide maximally exposed individual member of the public at the Livermore site and Site 300 from operations in 2012 are:

- Livermore site: 5.4×10^{-3} mrem (5.4×10^{-2} μ Sv)
- Site 300: 1.3×10^{-6} mrem (1.3×10^{-5} μ Sv)

Background Information

LLNL is a premier research laboratory that is part of the National Nuclear Security Administration (NNSA) within DOE. As a national security laboratory, LLNL is responsible for ensuring that the nation’s nuclear weapons remain safe, secure, and reliable. The Laboratory also meets other national security needs, including countering the proliferation of weapons of mass destruction and strengthening homeland security, and conducts major research in atmospheric, earth, and energy sciences; bioscience and biotechnology; and engineering, basic science, and advanced technology. The Laboratory serves as a scientific resource to the U.S. government and a partner to industry and academia.

Because LLNL is a DOE facility, it is subject to the requirements of 40 CFR 61, Subpart H, National Emission Standards for Emissions of Radionuclides Other Than Radon From Department of Energy Facilities. This regulation limits emissions of radionuclides to ambient air to levels resulting in an annual effective dose equivalent of 10 mrem (100 μ Sv) to any member of the public. The regulation also requires annual reporting of the emissions and resulting dose.

1.1 SITE DESCRIPTION

LLNL consists of two sites—an urban site in Livermore, California, referred to as the “Livermore site;” and a rural experimental test site, referred to as “Site 300,” near Tracy, California (**Figure 1**).



Figure 1. Locations of LLNL’s Livermore site and Site 300.

The Livermore site is just within the eastern city limits of Livermore, a city of about 82,000 in Alameda County. The site occupies 1.3 square miles, including the land that serves as a buffer zone around most of the site. Within a 50-mile radius of the Livermore site are communities

such as Tracy and Pleasanton and the more distant (and more densely populated) cities of Oakland, San Jose, and San Francisco. Of the 7.7 million people within 50 miles of the Laboratory, only about 10% are within 20 miles.

Site 300, LLNL's Experimental Test Site, is located in the Altamont Hills of the Diablo Range and straddles the San Joaquin and Alameda county line. The site is 12 miles east of the Livermore site and occupies 10.9 square miles. The city of Tracy, with a population of over 84,000, is approximately 6 miles to the northeast (measured from the northeastern border of Site 300 to Sutter Tracy Community Hospital). Of the 7.1 million people who live within 50 miles of Site 300, 95% are more than 20 miles away in distant metropolitan areas such as Oakland, San Jose, and Stockton.

The weather conditions at the Livermore site and Site 300 are very similar. The climate at both sites is best described as Mediterranean, characterized by mild, rainy winters and warm-to-hot, dry summers. However, the complex topography of Site 300 does influence local wind and temperature patterns. The stronger winds that occur at the higher elevations of Site 300 (see **Figure 2**), results in warmer nights and slightly cooler days than the Livermore site.

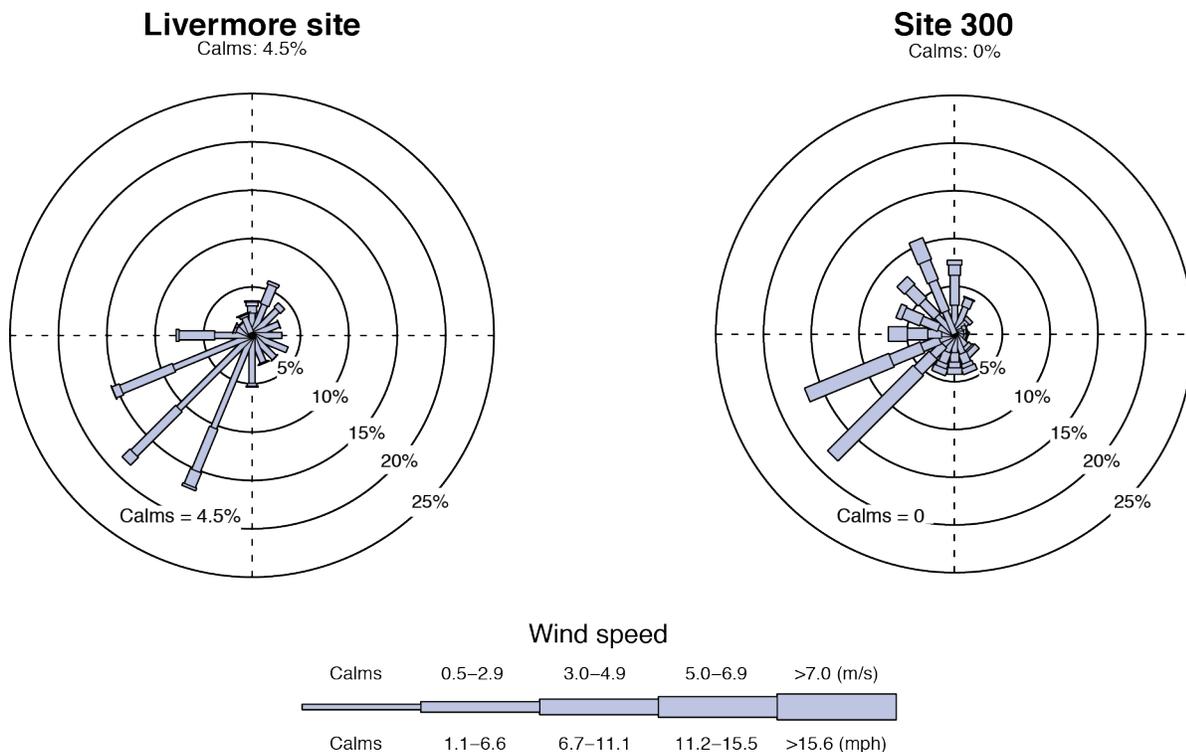


Figure 2. Wind roses for the Livermore site and Site 300 for 2012.

The 2012 annual wind data for both sites are displayed as wind roses in **Figure 2**. In the wind rose, the length of each spoke is proportional to the frequency at which the wind blows from the indicated direction; different line widths of each spoke represent wind speed classes. These data show that for the Livermore site, winds blew from the south-southwest through west-southwest

about 51% of the time; for Site 300, the data show that the winds blew from the southwest to the west-southwest about 35% of the time. The average wind speed in 2012 at the Livermore site was 2.3 m/s (5.1 mph), and the average wind speed at Site 300 was 5.6 m/s (12.6 mph). In 2012, the Livermore site received 30.6 cm of rain and Site 300 received 29.7 cm.

1.2 SOURCE DESCRIPTION

Many different radioisotopes were available for use at LLNL in 2012 for research purposes, including biomedical tracers, tritium, mixed fission products, transuranic isotopes, and others—see **Table 1**. Radioisotope handling procedures and work enclosures are determined for each project or activity, depending on the isotopes, the quantities being used, and the types of operations being performed. Work enclosures include glove boxes, exhaust hoods, and laboratory bench tops. Exhaust paths to the atmosphere include High Efficiency Particulate Air (HEPA) filtered ventilation systems, roof vents and stacks lacking abatement devices, resuspension of depleted uranium from the soil due to wind conditions and previous open-air explosives testing at Site 300, and releases to ambient air from a variety of diffuse sources.

Table 2 identifies the buildings, by managing organization, at LLNL where there was a potential for release of radioactive materials to the air in 2012.

Table 1. Radionuclides used at LLNL during 2012.

| | | | | | |
|---------|--------|--------|--------|--------|--------|
| Ag-110 | Cf-249 | Eu-155 | Ni-56 | Pu-239 | Th-228 |
| Ag-110m | Cf-252 | Fe-55 | Ni-57 | Pu-240 | Th-229 |
| Al-26 | Cl-36 | Fe-59 | Ni-59 | Pu-241 | Th-230 |
| Am-241 | Cm-243 | Fe-60 | Ni-63 | Pu-242 | Th-232 |
| Am-242m | Cm-244 | Gd-148 | Ni-66 | Pu-244 | Tl-204 |
| Am-243 | Cm-245 | H-3 | Np-236 | Ra-226 | U-232 |
| Ba-133 | Cm-246 | Hg-203 | Np-237 | Ru-105 | U-233 |
| Be-7 | Cm-248 | I-125 | Np-239 | Ru-106 | U-234 |
| Be-10 | Co-56 | I-129 | P-32 | S-35 | U-235 |
| Bi-207 | Co-57 | I-131 | P-33 | Sb-125 | U-236 |
| C-14 | Co-58 | Ir-192 | Pa-231 | Se-75 | U-238 |
| Ca-41 | Co-60 | K-40 | Pa-233 | Sr-85 | Xe-133 |
| Ca-45 | Cs-134 | Kr-85 | Pb-210 | Sr-89 | Xe-135 |
| Cd-109 | Cs-137 | Mn-54 | Po-210 | Sr-90 | Y-88 |
| Ce-139 | Eu-152 | Na-22 | Pu-236 | Tc-99 | Y-90 |
| Ce-141 | Eu-154 | Nb-95 | Pu-238 | Tc-99m | Zr-95 |
| Ce-144 | | | | | |

Table 2. Buildings at LLNL, by managing organization, where there is a potential for the release of radioactive materials to the air.

| Director's Office | Physical & Life Sciences | Engineering | Weapons & Complex Integration | National Ignition Facility & Photon Science | Operations & Business |
|--------------------------|-------------------------------------|--------------------|------------------------------------------|--------------------------------------------------------|----------------------------------|
| B253 | B132 ^a | B131 | B331 ^b | B162 | B419 |
| B254 | B151 | B231 | B332 ^b | B298 | B597 |
| B255 | B154 | B321 | B612 | B381 | |
| | B194 | B322 | B625 | B391 | |
| | B235 ^b | B327 | B695/696 ^b | B581 ^b | |
| | B282 | | B697 | B582 | |
| | B292 | | B801 ^b | B491 ^b | |
| | B341 | | B804 | | |
| | B361 | | B810A | | |
| | B364 | | B810B | | |
| | B378 | | B851 | | |
| | | | B883 | | |

^a B132 is managed by Global Security.

^b Continuous monitoring occurs at one or more exhaust points at the building.

Emissions Data

LLNL places radionuclide emission sources into one of two categories; major sources or minor sources. Major sources are defined as those that have the potential to emit radionuclides that could result in an annual potential dose of 0.1 mrem (1 μ Sv) or more to a member of the public at an off-site location; the radionuclide NESHAPs regulation requires continuous monitoring where the annual potential dose is in excess of 0.1 mrem (1 μ Sv). Minor sources are defined as sources that do not have the potential to cause an annual dose of 0.1 mrem (1 μ Sv). At LLNL, all major sources of emissions are point sources, i.e., stack emission points; however, minor sources include both point sources and diffuse sources.

2.1 MAJOR SOURCES: MEASURED EMISSIONS

In 2012, there were six facilities at the Livermore site and one facility at Site 300 that had radionuclide air effluent monitoring systems. These facilities are listed in **Table 3**, along with the number of samplers, the types of samplers, and the analytes of interest. Some of these facilities have the potential to emit radionuclides that would cause an annual dose in excess of the 0.1 mrem (1 μ Sv) standard; these sources are major sources following the definition given above. Others of these facilities have in the past had emissions required monitoring, and the monitoring has been maintained to assure that the emissions continue to be well characterized and that the potential effect on the public and the environment is well understood.

Many of the monitored stacks at LLNL have effluent controls, such as HEPA filters, to collect materials before they are emitted to the atmosphere. Air samples for particulate emissions are extracted downstream of HEPA filters and prior to the discharge point to the atmosphere. Particles are collected on cellulose membrane filters. The sample filters are removed and analyzed for radioactive particulate activity on a weekly or bi-weekly frequency depending on the facility. In all cases, continuous passive filter aerosol collection systems are used. At some facilities, continuous air monitors (CAMs) also sample the stack air exhaust for radionuclides. CAMs have an alarm capability for the facility in the event of an unplanned release of alpha activity. CAMs are used for facility personnel safety; they are not used for NESHAPs compliance demonstration

Table 3. Air effluent sampling systems and locations.

| Building | Facility | Analytes | Sample type | Number of samplers |
|----------|----------------------------------------------------|-----------------------------------------------------|---------------------------------|--------------------|
| 235 | Building in Physical and Life Sciences Directorate | Gross α , β on particles | Filter | 1 |
| 331 | Tritium Facility | Gaseous tritium/ tritiated water vapor | Ionization Chamber ^a | 4 |
| | | Gaseous tritium/ tritiated water vapor | Glycol Bubblers | 2 |
| 332 | Plutonium Facility | Gross α , β on particles | Filters | 15 |
| | | Gross α , β on particles | CAM ^a | 12 |
| 491 | Isotope Separation ^b | Gross α , β on particles | Filter | 1 |
| 581 | National Ignition Facility | Gross α , β , Gamma suite on particles | Filter | 1 |
| | | Radioiodine (volatile) | TEDA cartridge | 1 |
| | | Gaseous tritium/ tritiated water vapor | Glycol Bubbler | 1 |
| | | Gaseous tritium/ tritiated water vapor | Ionization Chamber ^a | 1 |
| 695/696 | Decontamination and Waste Treatment Facility | Gross α , β on particles | Filter | 1 |
| 801A | Contained Firing Facility (Site 300) | Gross α , β on particles | Filter | 1 |

Note: "CAM" denotes continuous air monitors.

^a Alarmed systems used for notification only so that any unplanned release may be detected and mitigated; they are not used for NESHAPs compliance demonstration.

^b Isotope separation operations are discontinued; area now used for storage of contaminated parts.

Detection of radioactive particulate activity resulting from particles collected on the air filters is accomplished using gas flow proportional counters and gamma spectroscopy.

For verification of the operation of the counting system, calibration sources, and background samples, are interspersed among with the sample filters for analysis. The Radiological Measurements Laboratory (RML) in LLNL's Radiation Protection Functional Area and the Environmental Monitoring Radio-analytical Laboratory (EMRL) in the Physical and Life Sciences Directorate perform the analyses.

When the result for gross alpha or gross beta on a particulate sample is greater than the minimum detectable concentration (MDC) for gross alpha activity, the filter is recounted. If the second result is also above the MDC, the filter is submitted to the EMRL for isotopic analysis to determine whether the activity on the filter is the result of naturally occurring radionuclides or is reportable as a radionuclide emission from the facility.

Glycol bubblers are used to monitor for tritium releases from the two Tritium Facility (Building 331) stacks, and the National Ignition Facility (NIF) stack. In addition to this

NESHAPs compliance monitoring, the two Tritium Facility stacks, and the NIF stack are monitored using ion chambers. The ion chamber monitors are set to alarm at designated tritium concentrations to identify accidental or off-normal releases. Ion chambers are in place for notification only so that any unplanned release may be detected and mitigated; they are not used for NESHAPs compliance demonstration, but data may serve as supportive information. All of the stack samplers monitor continuously.

Because tritium can be released in the form of either tritiated water vapor (HTO) or gaseous tritium (HT), glycol bubblers employ a two-stage glycol impinging process to capture each physical form. Stack air to be sampled enters the instrument and flows through the first stage impingers, capturing the HTO present. Next, the sampled air is directed through a heated palladium catalyst where oxidation of any HT in the sample takes place, converting gaseous tritium to HTO, which is then collected in the second stage impingers. The impingers are analyzed by the RML using liquid scintillation analysis. This type of sampling quantifies the amount of tritium for both species, HT and HTO.

Tritium in particulate form is monitored when appropriate with the use of cellulose membrane filters. Measurements to date indicate that tritium exchange (adsorption of tritium in HTO and/or HT is captured in the filter medium via a binding reaction; this is verified by placing two particulate filters in series and getting equal results with 2-sigma error applied) accounts for the tritiated particulate detections to date.

Triethylenediamine (TEDA) cartridges are used to sample for radioactive iodines in gaseous or vapor state. The TEDA is impregnated into carbon (activated carbon) by the manufacturer and is housed in a plastic cartridge of standard industry size 2 1/4" diameter by 1" thick (30 × 50 Mesh). Stack air is directed through the TEDA cartridge that is located post a particulate filter (two-stage filter housing). Both the particulate filter and the TEDA cartridge are counted by gamma spectroscopy by the EMRL.

In 2012, a total of 51.3 Ci (1898 GBq) of measured tritium was released from the Tritium Facility. Of this, approximately 88% of tritium was released as vapor (HTO). The remaining 12% released was gaseous tritium (HT).

The National Ignition Facility (NIF) released a total of 8.51 Ci (315 GBq) of measured tritium from the stack exhaust in 2012. A total of 1.31 Ci (48.5 GBq) was released as vapor (HTO) and 7.21 Ci (267 GBq) as gaseous tritium (HT).

The Contained Firing Facility at Site 300 had measured depleted uranium emissions in 2012. A total of 4.3×10^{-8} Ci (1.6×10^{-6} GBq) of uranium-234, 3.6×10^{-9} Ci (1.3×10^{-7} GBq) of uranium-235, and 3.4×10^{-7} Ci (1.3×10^{-5} GBq) of uranium-238 was released in particulate form.

None of the other facilities monitored for radionuclides had reportable emissions in 2012.

2.2 MINOR SOURCES: AMBIENT MEASUREMENT COMPARISON

With EPA's Region IX approval, LLNL demonstrates compliance for minor emissions sources (both non-monitored stack and area sources) through the use of ambient air monitoring data. The method entails comparing measured ambient air concentrations at the location of the site-wide maximally exposed individual (SW-MEI) to concentration limits set by EPA in its Table 2 of Appendix E to 40 CFR 61. The radionuclides for which the comparisons are made are tritium and plutonium 239+240 for the Livermore SW-MEI and uranium-238 for the Site 300 SW-MEI (see **Table 6** in Section 3.3.2).

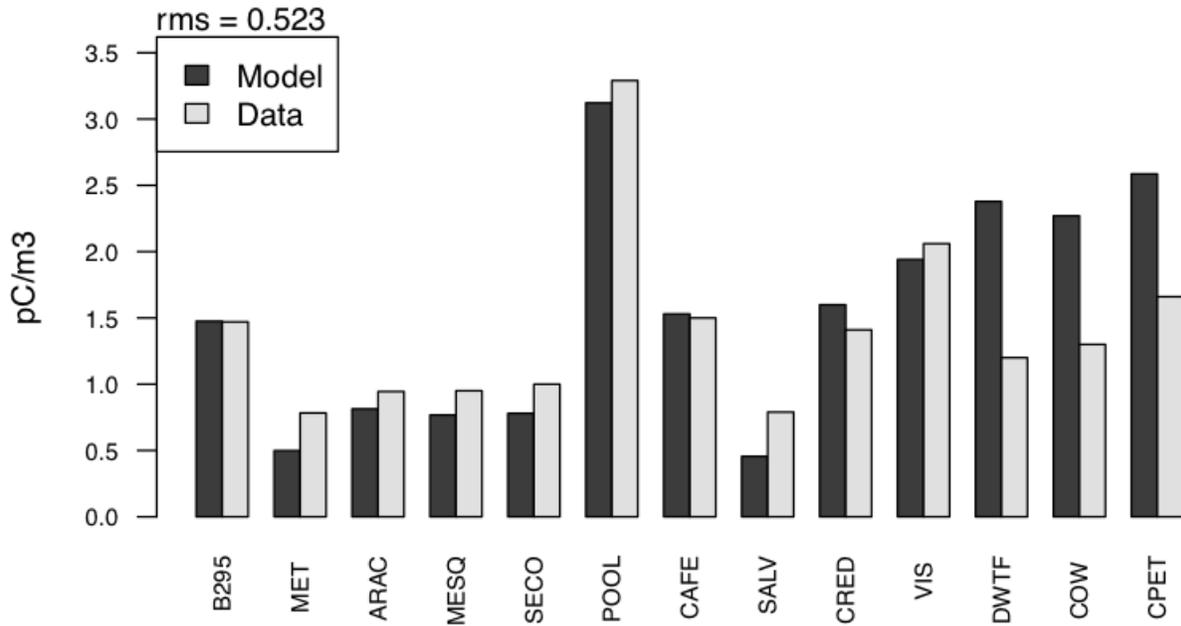
2.3 MINOR SOURCES: SOURCE TERM ESTIMATE

In order to take into account the dose contributions from diffuse minor sources, LLNL estimated the source terms for these sources using a mathematical optimization routine that minimizes the root-mean-square (*rms*) differences between modeled and measured average annual ambient tritium concentrations (see **Figure 5** in Section 3.3.2 for the ambient air sampling locations). This process has two parts.

First, CAP88-PC is used to model the contribution each source (both point and diffuse) makes to the annual average tritiated water vapor (HTO) concentration at each of the ambient air monitoring locations. Each point source is modeled using source-specific model parameters (activity, stack height, etc.). The diffuse sources are modeled using a unit source (1 Ci), a 1-meter height, a 10-meter diameter, and a fixed plume rise across stability classes A through F. All models use the same LLNL 2012 wind file. The individual contributions (both point and diffuse) at each monitoring location are added to produce an all-sources-combined model estimate of the annual average ambient concentration at each location.

Second, the 1 Ci source term for each diffuse source is adjusted independently using a mathematical search optimization routine. The point sources are held fixed. The adjusted diffuse source terms that give the best fit of the all-sources-combined model concentrations to the measured concentrations are then used to calculate the dose contribution from each diffuse source.

In 2012, the best-fit diffuse source term estimates are 1.3 Ci for the Building 331 Waste Accumulation Area (WAA), 0.12 Ci for Building 298, and 0.22 Ci for the Building 251 Yard. These estimates are consistent with the reported activities and contents of those areas. The measured and best-fit model annual average results are shown in **Figure 3**.



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Figure 3. Comparison of measured and modeled annual average concentrations of tritiated water vapor (HTO) in units of pCi/m³ in air at Livermore site ambient air locations, 2012.

2.4 MINOR SOURCES: OPEN-AIR TESTS

Another potential source of radioactive air emissions from LLNL operations at Site 300 is the emission of materials from open-air explosives tests. In 2012, there were no open-air explosives tests that contained radioactive materials.

Dose Assessment

3.1 GENERAL

To comply with NESHAPs regulations and DOE guidance, the EPA-approved atmospheric dispersion and radiation dose calculation computer code, CAP88-PC, Version 1.0, was used to calculate the dose at various distances and from various release points. For diffuse sources having a significant contribution to total dose, in addition to comparing the emissions to the concentration limits set by EPA in its Table 2 of Appendix E to 40 CFR 61, doses were calculated using either CAP88-PC or standard breathing rates and dose conversion factors.

For LLNL to comply with the NESHAPs regulations, the LLNL SW-MEI cannot receive an effective dose equivalent greater than 10 mrem/y ($100 \mu\text{Sv/y}$). The SW-MEI is defined as the *hypothetical* member of the public at a single residence, school, business, church, or other such facility who receives the greatest LLNL- induced dose from the combination of all evaluated radionuclide source emissions, as determined by modeling. At the Livermore site, the SW-MEI for 2012 was located at the UNCLE Credit Union, about 30 feet (10 m) outside the controlled eastern fence line of the site, but about 30 feet (10 m) within the perimeter of the site property. At Site 300, the 2012 SW-MEI was located at the boundary with the Carnegie State Vehicle Recreation Area, managed by the California Department of Parks and Recreation, approximately 1.9 miles (3.2 km) south-southeast of the firing table at Building 851. The locations of the SW-MEIs for both LLNL sites are shown in **Figure 4**.

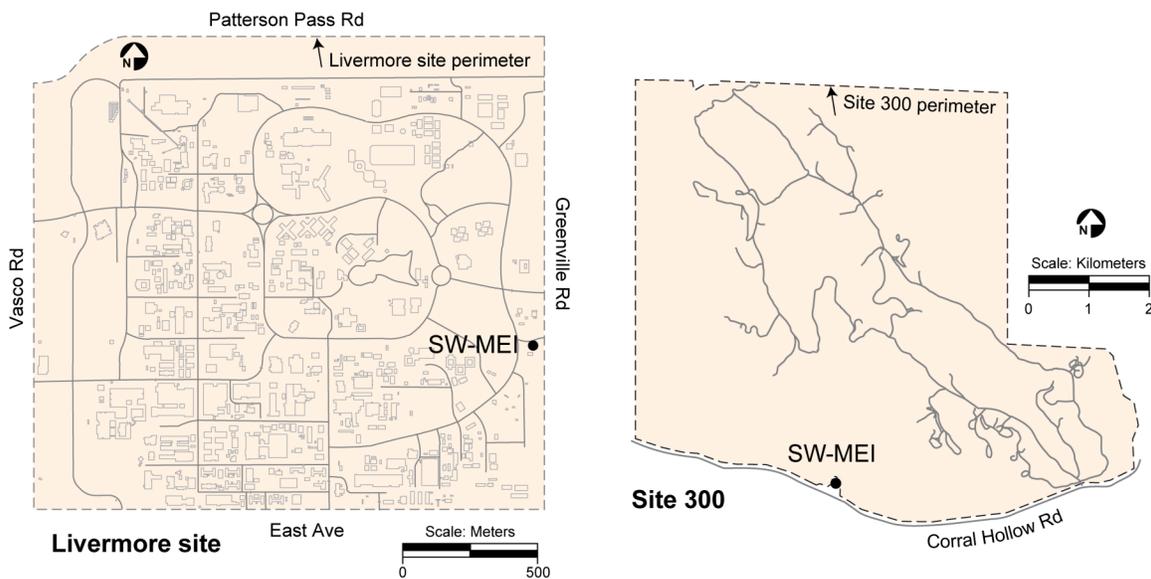


Figure 4. Location of Site-Wide Maximally Exposed Individual (SW-MEI) at the Livermore site and Site 300, 2012.

3.2 CAP88-PC INPUT PARAMETERS

Input parameters to CAP88-PC include the emissions discussed in Section 2, and building-specific and common parameters, discussed below. To estimate dose, CAP88-PC, Version 1, provides a library of 265 radionuclides. In addition, when calculating dose from particulate alpha- and beta-emitting radionuclides, LLNL assigns gross alpha and gross beta measurements to the radionuclides handled in the facility, when they can be specifically identified, or to plutonium-239+240 and strontium-90, respectively. The use of plutonium-239+240 and strontium-90 to represent alpha and beta emissions provides a health-conservative estimate of the dose.

3.2.1 Building-Specific Parameters

For dose assessment, LLNL uses building-specific information about radionuclide releases, as well as building-specific parameters for stack height, stack exhaust rate, stack diameter, and distances to the fence line. The building specific parameters are presented in **Attachment 1**.

3.2.2 Common Parameters

The input parameters that are common among LLNL sources are the agricultural parameters. Meteorological data from the LLNL Livermore site meteorological tower are used to model Livermore site sources, and meteorological data from the LLNL Site 300 meteorological tower are used to model Site 300 sources. Site-specific values for annual precipitation (12.0 in. [30.6 cm] for the Livermore site and 11.7 in. [29.7 cm] for Site 300) and annual average ambient temperature (58.6°F [14.8°C] for the Livermore site and 62.6°F [17.0°C] for Site 300) were used. The CAP88-PC, Version 1, default for absolute humidity, 8 g/m³, is reasonably representative of conditions at LLNL, was used. The value for lid (mixing) height of 2,460 ft (750 m) was chosen for the Livermore site, whereas the lid height value for Site 300 was 3,280 ft (1,000 m). The 2012 wind data are provided in **Attachment 2**.

For agricultural parameters in CAP88-PC, LLNL used mean values for California based on data from the U.S. Department of Agriculture (USDA 2007). The mean values are shown in **Table 4**.

Table 4. Agricultural parameter values representing LLNL used in CAP88-PC.

| Parameter | Value |
|-----------------------------------------------|-------|
| Beef cattle density (# cows/km ²) | 4.8 |
| Milk cattle density (# cows/km ²) | 0.025 |
| Land fraction cultivated for vegetable crops | 0.065 |

For individual and collective doses from ingestion, it was assumed that 100% of milk is imported (i.e., free from LLNL-generated radioactivity), and that vegetables and meat are 25% home-grown and 75% imported.

3.3 COMPLIANCE ASSESSMENT

3.3.1 Major Sources

Doses from LLNL's major sources, which are point sources for which monitoring is required, were evaluated using CAP88-PC and the input parameters discussed above. The modeled doses to the SW-MEI for the facilities where there were measurements greater than the minimum detectable concentration (MDC) are shown in **Table 5**. The specific results for all sources are provided in **Attachment 1**.

Table 5. Point source doses for 2012.

| Facility | Dose (mrem) |
|-------------------------|----------------------|
| Tritium Facility (B331) | 4.8×10^{-3} |
| NIF (B581) | 1.5×10^{-4} |
| CFF (B801A) | 1.3×10^{-6} |

3.3.2 Minor Sources

LLNL has many minor sources; most of them are point sources and a few are diffuse. As stated previously, with EPA's Region IX approval, LLNL demonstrates compliance for minor emissions sources (both diffuse and non-monitored stack sources) through the comparison of ambient air monitoring data with concentration limits set by EPA in Table 2 of Appendix E to 40 CFR 61. This is done for tritium and plutonium-239+240 for the Livermore SW-MEI and uranium-238 for the Site 300 SW-MEI. The 2012 average monitoring results for tritium and plutonium from the sampling location in closest proximity to the Livermore site SW-MEI (UNCLE Credit Union [CRED]) were used to represent the SW-MEI for the purposes of this minor source comparison. (See **Figure 5** for a map of all Livermore site sampling locations).

The results of these comparisons are shown in **Table 6**. In 2012, all monthly measurements for plutonium-239+240 at CRED were non-detections. At Site 300, wind-driven resuspension of soil contaminated with depleted uranium is of greatest interest in the minor source category. Since 2008, but in contrast to years prior, no ambient measurements for uranium showed a contribution from depleted uranium—the uranium-238 value in **Table 6** represents a natural background value (see footnote c). The lack of measurements indicating the presence of depleted uranium at the SW-MEI is related to no outdoor explosives tests in 2012 that included depleted uranium. Because there was no source term for depleted uranium resuspension at Site 300, there is no minor source contribution to the calculated dose for 2012 at the SW-MEI.

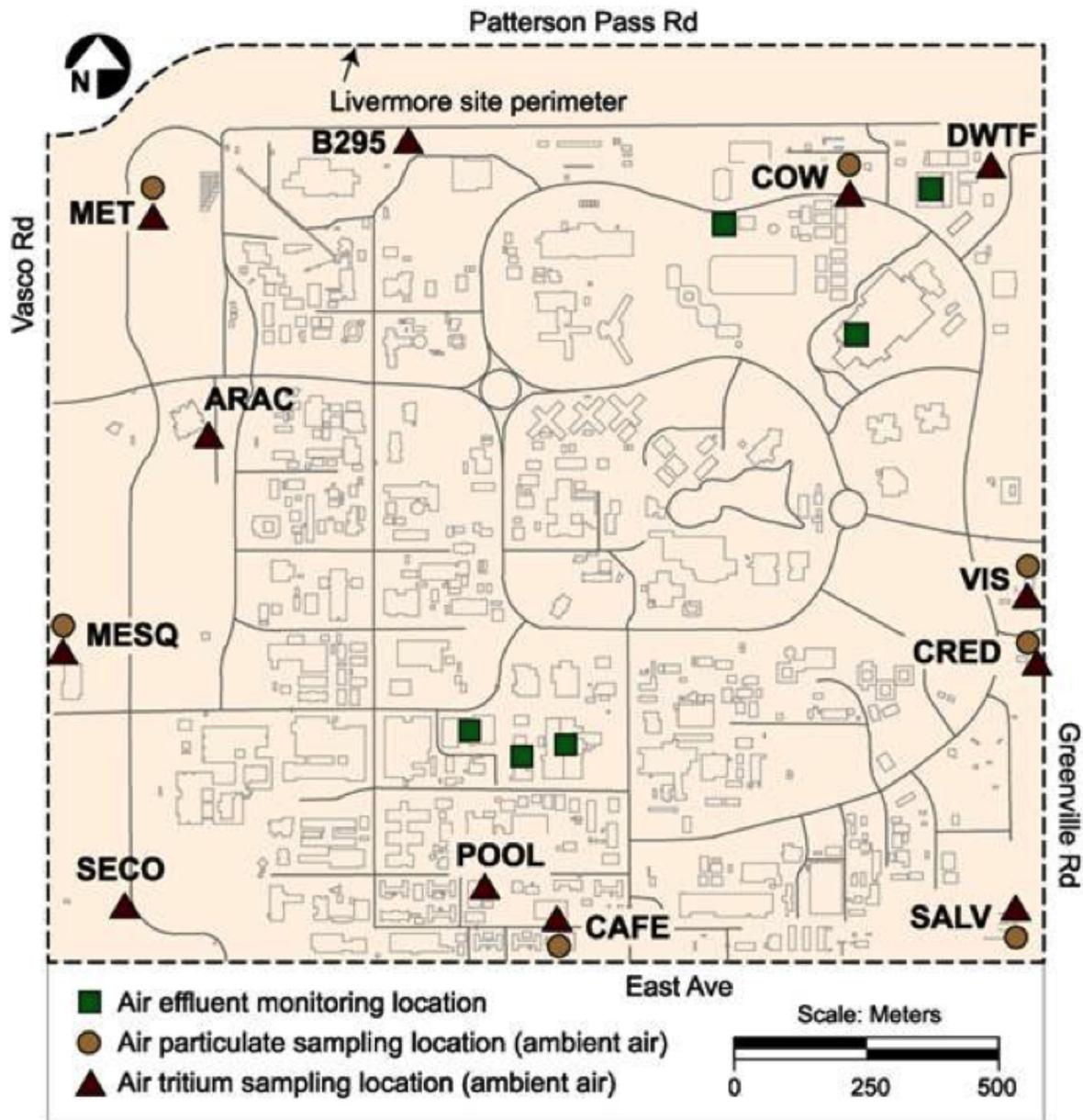


Figure 5. Radiological air monitoring locations at the Livermore site in 2011.

The measured concentrations at the SW-MEI are presented in **Table 6**. Also shown in **Table 6** are EPA's standards from Table 2 of Appendix E to 40 CFR 61. As demonstrated by the calculation of the fraction of the standard, LLNL's measured concentrations in air for tritium, plutonium-239+240, and uranium-238 are a fraction less than 0.002 (0.2%) of the standard for these radionuclides.

Table 6. Mean concentrations of radionuclides of concern at the location of the SW-MEI in 2012 compared to EPA's concentration standard.

| Location | Nuclide | EPA's Table 2 concentration standard | Mean measured concentration | Measured concentration as a fraction of the standard | Detection limit |
|-----------------------|---------------|-----------------------------------------|------------------------------------------|------------------------------------------------------|---------------------------------------|
| Livermore site SW-MEI | Tritium | 1.5×10^{-9} Ci/m ³ | 1.4×10^{-12} Ci/m ^{3a} | 9.3×10^{-4} | 1×10^{-12} Ci/m ³ |
| Livermore site SW-MEI | Plutonium-239 | 2.0×10^{-15} Ci/m ³ | Non-detect ^b | Non-detect ^b | 5×10^{-19} Ci/m ³ |
| Site 300 SW-MEI | Uranium-238 | 8.3×10^{-15} Ci/m ³ | 1.6×10^{-17} Ci/m ^{3c} | 1.9×10^{-3} | 3×10^{-20} Ci/m ³ |

^a The measured tritium value includes contributions from all major and minor sources including stack and diffuse releases at the location of the SW-MEI.

^b All results for Pu-239 at the SW-MEI location were below the analytical detection limits.

^c The average ratio of uranium-238 and uranium-235 concentrations for 2012 is 0.0072, which is the ratio of these isotopes for naturally occurring uranium. This value for uranium-238 is from naturally occurring uranium resuspended to ambient air from the soil.

The source term for diffuse sources of tritium was developed using a mathematical model (Section 2.3) and the doses were calculated using CAP88-PC (**Attachment 1** lists the doses from diffuse sources). The total diffuse source dose for 2012 was 4.1×10^{-4} mrem (4.1×10^{-3} μ Sv) for the Livermore site; because there was no diffuse source term for Site 300, no minor source diffuse dose was calculated.

3.3.3 MEI Dose

Doses from LLNL's airborne emissions are well below the 10 mrem (100 μ Sv) NESHAPs annual dose standard. The annual doses to the hypothetical SW-MEI at the Livermore site and at Site 300 are:

- Livermore site: 5.4×10^{-3} mrem (5.4×10^{-2} μ Sv)
- Site 300: 1.3×10^{-6} mrem (1.3×10^{-5} μ Sv)

The EPA-approved software calculates the dose assuming a person resides there all year for 24 hours a day, eats meat and vegetables grown at the location (see agricultural parameters in Section 3.2.2), and drinks contaminated water. Thus, the calculated dose to this hypothetical person, the SW-MEI, is greater than the dose to an actual resident.

Table 7 presents 2012 doses with those of previous years. Diffuse source doses were not reported for the Livermore site for 1990 and 1991, and were not reported for Site 300 for 1990 through 1992.

Table 7. Doses (in mrem) calculated for the Site-Wide Maximally Exposed Individual (SW-MEI) for the Livermore site and Site 300, 1990 to 2012.

| Year | Total Dose | Point Source Dose | Diffuse Source Dose |
|-----------------------|------------------------|------------------------|---------------------|
| Livermore site | | | |
| 2012 | 0.0054 | 0.005 | 0.00041 |
| 2011 | 0.017 | 0.015 ^a | 0.0019 |
| 2010 | 0.011 | 0.0033 ^a | 0.0074 |
| 2009 | 0.0042 ^a | 0.0015 ^a | 0.0027 |
| 2008 | 0.0013 ^a | 0.00033 ^a | 0.00095 |
| 2007 | 0.0031 ^a | 0.0013 ^a | 0.0018 |
| 2006 | 0.0045 ^a | 0.0016 ^a | 0.0029 |
| 2005 | 0.0065 ^a | 0.0027 ^a | 0.0038 |
| 2004 | 0.0079 ^a | 0.0021 ^a | 0.0058 |
| 2003 | 0.044 ^a | 0.024 ^a | 0.020 |
| 2002 | 0.023 ^a | 0.010 ^a | 0.013 |
| 2001 | 0.017 ^a | 0.0057 ^a | 0.011 |
| 2000 | 0.038 ^a | 0.017 ^a | 0.021 |
| 1999 | 0.12 ^a | 0.094 ^a | 0.028 |
| 1998 | 0.055 ^a | 0.031 ^a | 0.024 |
| 1997 | 0.097 | 0.078 | 0.019 |
| 1996 | 0.093 | 0.048 | 0.045 |
| 1995 | 0.041 | 0.019 | 0.022 |
| 1994 | 0.065 | 0.042 | 0.023 |
| 1993 | 0.066 | 0.040 | 0.026 |
| 1992 | 0.079 | 0.069 | 0.010 |
| 1991 | 0.234 | — ^b | — ^b |
| 1990 | 0.240 | — ^b | — ^b |
| Site 300 | | | |
| 2012 | 1.3 × 10 ⁻⁶ | 1.3 × 10 ⁻⁶ | — ^c |
| 2011 | 9.0 × 10 ⁻⁸ | 9.0 × 10 ⁻⁸ | — ^c |
| 2010 | 5.7 × 10 ⁻⁷ | 5.7 × 10 ⁻⁷ | — ^c |
| 2009 | 2.7 × 10 ⁻⁷ | 2.7 × 10 ⁻⁷ | — ^c |
| 2008 | 4.4 × 10 ⁻⁸ | 4.4 × 10 ⁻⁸ | — ^c |
| 2007 | 0.0035 | 0.0031 | 0.00035 |
| 2006 | 0.016 | 0.014 | 0.0020 |
| 2005 | 0.018 | 0.0088 | 0.0094 |
| 2004 | 0.026 | 0.025 | 0.00086 |
| 2003 | 0.017 | 0.017 | 0.00034 |
| 2002 | 0.021 | 0.018 | 0.0033 |
| 2001 | 0.054 | 0.050 | 0.0037 |
| 2000 | 0.019 | 0.015 | 0.0037 |
| 1999 | 0.035 | 0.034 | 0.0012 |
| 1998 | 0.024 | 0.019 | 0.005 |
| 1997 | 0.020 | 0.011 | 0.0088 |
| 1996 | 0.033 | 0.033 | 0.00045 |
| 1995 | 0.023 | 0.020 | 0.003 |
| 1994 | 0.081 | 0.049 | 0.032 |
| 1993 | 0.037 | 0.011 | 0.026 |
| 1992 | 0.021 | 0.021 | — ^d |
| 1991 | 0.044 | 0.044 | — ^d |
| 1990 | 0.057 | 0.057 | — ^d |

^a The dose includes HT emissions modeled as HTO. Modeling HT emissions as such results in an overestimation of the dose. This methodology is used for purposes of compliance, as directed by EPA Region IX.

^b Point and diffuse source doses were not reported separately from the total dose for the Livermore site for 1990 and 1991.

^c No diffuse emissions dose was calculated for 2008, 2009, 2010, 2011, and 2012 because ambient monitoring yielded no results indicating the presence of depleted uranium at the SW-MEI.

^d No diffuse emissions were evaluated at Site 300 for years before 1993.

Certification

I certify under penalty of law that this document and all attachments were prepared under my direction or supervision in accordance with a system designed to assure that qualified personnel properly gather and evaluate the information submitted. Based on my inquiry of the person or persons who manage the system, or those persons directly responsible for gathering the information, the information submitted is, to the best of my knowledge and belief, true, accurate, and complete. I am aware that there are significant penalties for submitting false information, including the possibility of fine and imprisonment for knowing violations.

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Frances Alston

I certify under penalty of law that I have personally examined and am familiar with the information submitted herein, and based on my inquiry of those individuals immediately responsible for obtaining the information, I believe that the submitted information is true, accurate, and complete. I am aware that there are significant penalties for submitting false information, including the possibility of fine and imprisonment. See 18 U.S.C. 1001.

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Additional Information

5.1 UNPLANNED RELEASES

There were no unplanned releases from the Livermore site or Site 300 in 2012.

Supplemental Information

6.1 COLLECTIVE DOSE ASSESSMENT

Collective population dose is calculated using CAP88-PC as the average radiation dose to a person in a specified area, multiplied by the number of people in that area. In accordance with DOE and EPA guidance documents, all radionuclides potentially emitted in 2012 were assumed to be released from a central location. The total population within 50 miles (80 km) of the Livermore site is approximately 7,770,000, and the total population within 50 miles (80 km) of Site 300 is approximately 7,110,000. The populations were derived using ORNL LANDSCAN™ 2010 data and ESRI ARCMAP software. The population file is provided in **Attachment 3**. The estimated collective dose attributable to LLNL airborne emissions in 2012 to persons living within 50 miles (80 km) of the Livermore site is 0.68 person-rem (0.0068 person-Sv) and to persons living within 50 miles (80 km) of Site 300 is 2.3×10^{-4} person-rem (2.3×10^{-6} person-Sv).

6.2 40 CFR 61 SUBPARTS Q AND T

LLNL does not have storage and disposal facilities for radium containing materials that would be a significant source of radon. Emissions of radon from LLNL research experiments did not occur in 2012. LLNL does not have or store any uranium mill tailings.

6.3 PERIODIC CONFIRMATORY MEASUREMENT

Results of NESHAPs periodic confirmatory measurements (PCM) are intended to support or confirm two objectives: 1) that those operations not continuously monitored do not, in fact, need to be continuously monitored and 2) that radionuclide usage-inventory-based estimates of emissions and their corresponding doses are conservative.

For sources evaluated to have a potential to result in a dose less than the regulatory value of 0.1 mrem/y that requires continuous monitoring under Subpart H, LLNL achieves the PCM objectives by fulfilling the requirements stated in 40 CFR 61.93, paragraph (e) with its ambient air monitoring program. The ambient air monitoring effort includes thirty-two sampling locations with forty-six samplers placed in strategic areas (see the Air Monitoring Programs section in the LLNL Site Annual Environmental Report [<https://saer.llnl.gov/>] for a description of LLNL's ambient air radiological monitoring).

6.4 FACILITY COMPLIANCE

In 2012, LLNL maintained its compliance with 40 CFR 61 Subpart H. All emissions resulted in calculated doses well below the 10 mrem (100 μ Sv) standard. **Attachment 1** provides the dose estimates for each individual source.

References

LandScan™ Global Population Database, 2010,
http://www.ornl.gov/sci/landscan/landscan_documentation.shtml

EPA 1989: U.S. Environmental Protection Agency, National Emission Standard for Emissions of Radionuclides Other Than Radon From Department of Energy Facilities, 40 CFR Part 61, Subpart H (1989, as amended).

National Council on Radiation Protection and Measurements (NCRP), Principles and Application of Collective Dose in Radiation Protection, NCRP Report No. 121 (1995).

USDA 2007. United States Department of Agriculture. The Census of Agriculture. 2007 Census Publications. Volume 1, Chapter 2, County Level Data. Table 1 and Table 11.
http://www.agcensus.usda.gov/Publications/2007/Full_Report/Volume_1,_Chapter_2_County_Level/California/index.asp (accessed May 19, 2009).

Errata

In the 2011 NESHAPs report, Attachment 1 spreadsheet, the B581 stack diameter is stated as 1.1 meters; it is 1.3 meters in diameter.

Attachments

A-1 through A-3

Attachment 1 - 2012 LLNL NESHAPs Annual Report Spreadsheet

| Building | Room/Area | Stack ID | Operation | Radionuclides | Monitoring for Potential of Release | Stack Height (m) | Stack Diameter (m) | Stack Velocity (m/s) | Control Device(s) | Control Device Abatement Factor | Estimated Annual Emissions (Ci) | 10 mrem/y Site-Wide Dose Requirement | | | 0.1 mrem/y Monitoring Requirement | | |
|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------------|-----------------------------------------------------------|--------------------------------------------------------------|---------------------------------------------------------------|------------------------------------------------------------------------------|------------------|------------------------|----------------------|-------------------------------------------------------------------------------|---------------------------------|------------------------------------------------------|--------------------------------------|---------------------|-----------------------------------------------------|-----------------------------------|------------------|-----------------------------------------------------------------------------------------|
| | | | | | | | | | | | | Distance to SW-MEI (m) | Direction to SW-MEI | EDE (mrem) | Distance to MEI (m) | Direction to MEI | EDE (mrem) |
| LIVERMORE SITE POINT SOURCES | | | | | | | | | | | | | | | | | |
| Building 235 is part of the Physical and Life Sciences Directorate. Operations in the facility include examination of material structure, surface, and subsurface; precision cutting, ion implanting, and metallurgical studies. | | | | | | | | | | | | | | | | | |
| 235 | 1130 | FHE-1A/1B, FHE2A/2B, and FGBE-1A/1B through FHE-1000/2002 | Preparation of plutonium samples for diamond anvil studies | Gross alpha Gross beta | * | 10.7 | 0.30 | 6.0 | Double HEPA | 0.0001 | 0.0E+00 0.0E+00 | 1065 | ENE | 0.0E+00 | b | b | b |
| Building 331 is operated by the Weapons and Complex Integration (WCI) Directorate. The building houses the tritium research facility and associated laboratories. | | | | | | | | | | | | | | | | | |
| 331 | All | Stack 1 Stack 2 | Tritium research and development Decontamination of parts | H-3 H-3 | ^a ^a | 30.0 30.0 | 1.22 1.22 | 6.7 10.3 | None None | 1 1 | 2.37E+00 4.89E+01 | 957 | ENE | 4.8E-03 | 957 | ENE | 4.8E-03 |
| Building 332 is operated by the WCI Directorate for plutonium research. Exhausts from glove box operations and the workplace are double or triple filtered by high efficiency particulate air (HEPA) filters. Exhausts are monitored with both continuous filter sampling and plutonium-specific, continuous real-time monitors (CAMs). | | | | | | | | | | | | | | | | | |
| 332 | Increment 1 Rooms | FHE-1000/2000 | Plutonium research | Transuranics | * | 8.8 | 0.8 x 1.1 | 17.3 | Double HEPA | 0.0001 | 0.0E+00 | 912 | ENE | 0.0E+00 | b | b | b |
| 332 | Increment 1 Glove boxes | FGBE-1000/2000 | Plutonium research | Transuranics | * | 11 | 0.3 | 5.4 | Triple HEPA | 0.000001 | 0.0E+00 | 912 | ENE | 0.0E+00 | b | b | b |
| 332 | Loft | FE-4.5W FE-4.5E | Loft exhaust Loft exhaust | Transuranics Transuranics | ^a ^a | 11 11 | 0.6 x 0.9 0.6 x 0.9 | 4.7 4.1 | HEPA HEPA | 0.01 0.01 | 0.0E+00 0.0E+00 | 912 912 | ENE ENE | 0.0E+00 0.0E+00 | b b | b b | b b |
| 332 | Increment 1 Glove boxes | FGBE-3000/4000 | Plutonium research | Transuranics | * | 11 | 0.3 | 5.3 | Triple HEPA | 0.000001 | 0.0E+00 | 912 | ENE | 0.0E+00 | b | b | b |
| 332 | Increment 3 Room and Glove boxes | FFE-1000/2000 FGBE-7000/8000 | Plutonium research Plutonium research | Transuranics Transuranics | ^a ^a | 10.1 10.1 | 0.9 0.27 | 11.1 2.6 | Room—Double HEPA Glove Box—Triple HEPA | 0.0001 0.000001 | 0.0E+00 0.0E+00 | 912 | ENE | 0.0E+00 0.0E+00 | b b | b b | b b |
| Building 491 is operated by the National Ignition Facility and Photon Science as an area for the storage of contaminated parts and classified laser research. Isotope separation activities that previously occurred in this building have been discontinued. The facility operates with two in-series high efficiency particulate (HEPA) filter banks to control emissions. | | | | | | | | | | | | | | | | | |
| 491 | All | FFE-1 | Storage | Gross alpha Gross beta | ^{aa} ^{aa} | 9.1 | 0.9 | 3.6 | Double HEPA | 0.0001 | 0.0E+00 0.0E+00 | 1000 | SSE | 0.0E+00 0.0E+00 | b | b | b |
| Building 581 is operated by the National Ignition Facility and Photon Science Directorate. Operations of the facility include inertial confinement fusion experiments and laser related research. Stack exhaust is abated with HEPA filters, activated carbon filters, and molecular sieves. The stack exhaust is continuously monitored for radionuclides. | | | | | | | | | | | | | | | | | |
| 581 | NIF | FE-1 | ICF Research | Gross alpha Gross beta Gamma Tritium Radioiodines | ^a ^a ^c ^d ^e | 35 | 1.3 | 11.7 | Double HEPA Double Molecular Sieves Double Activated Carbon Filters | 0.0001 0.01 0.01 | 0.0E+00 0.0E+00 0.0E+00 8.51E+00 0.0E+00 | 705 | SSE | 0.0E+00 0.0E+00 0.0E+00 1.5E-04 0.0E+00 | 336 | ENE | ^{bb} ^{bb} ^{bb} 1.5E-3 ^b ^{bb} |
| Building 695/696 is the Decontamination Waste Treatment Facility operated by Radiological and Hazardous Waste Management in WCI. All operations are HEPA filtered and have pre-filters in place; some operations have additional HEPA filtration. | | | | | | | | | | | | | | | | | |
| 695/696 | DWTF | FHE 1000/2000/3000 | Waste treatment | Gross alpha Gross beta | ^a ^a | 20.0 | 1.98 | 11.0 | HEPA Pre-filter | 0.01 0.1 | 0.0E+00 0.0E+00 | 953 | S | 0.0E+00 0.0E+00 | 198 | ENE | ^{bb} ^{bb} |
| SITE 300 POINT SOURCES | | | | | | | | | | | | | | | | | |
| Building 801 is the Contained Firing Facility, where explosives tests are conducted. This facility and the 851 Firing Table are operated by the Weapons and Complex Integration Directorate. | | | | | | | | | | | | | | | | | |
| 801 | Contained Firing Facility | FEFH-1, FE-2 | Explosive tests | U-238 U-235 U-234 | ^a ^a ^a | 16.8 | 1.60 | 5.1 | HEPA Pre-filter | 0.01 0.1 | 3.4E-07 3.6E-09 4.3E-08 | 3770 | S | 1.3E-06 | 1809 | ENE | 7.5E-6 ^b |
| Explosives tests in which radionuclides may be present are conducted on open-air firing tables located at Bunker 851. There were no atmospheric tests using depleted uranium or any other radioactive material in 2012. | | | | | | | | | | | | | | | | | |
| 851 | Firing Table | None | Explosive tests | U-238 U-235 U-234 | ^c ^c ^c | NA | NA | NA | None | 1 | 0.0E+00 0.0E+00 0.0E+00 | 3170 | SSE | 0.0E+00 | N/A | N/A | N/A |
| LIVERMORE SITE DIFFUSE SOURCES | | | | | | | | | | | | | | | | | |
| Building 331 - Contaminated equipment outside the facility awaiting decontamination or transport and storage by Radioactive and Hazardous Waste Management. | | | | | | | | | | | | | | | | | |
| 331 | Outside | None | Storage of contaminated parts | Tritium | ^f | NA | NA | NA | None | 1 | 1.3E+00 | 959 | ENE | 3.8E-04 | 445 | SSW | 1.2E-03 |
| Building 298 is operated by the National Ignition Facility where Tritium Target fabrications occur. | | | | | | | | | | | | | | | | | |
| 298 | Outside | Area Source | Storage of low level waste | Tritium | ^f | NA | NA | NA | None | 1 | 1.2E-01 | 1390 | SE | 4.7E-06 | 260 | NNE | 7.9E-04 |
| Building 251 outside yard was used to stage containers of tritium watch dials planned for waste recovery. The containers can outgas tritium. | | | | | | | | | | | | | | | | | |
| 251 | Outside | Area Source | Storage of low level waste | Tritium | ^f | NA | NA | NA | None | 1 | 2.2E-01 | 1151 | E | 2.1E-05 | 1168 | NNE | 9.3E-05 |
| The Southeast Quadrant of the Livermore Site has slightly elevated levels of Pu-239 in the surface soil and air. The source of the Pu-239 was past waste management operations. The CY12 results for PU resuspension at the location of the SW-MEI (SE Quadrant) were below analytical detection limits. | | | | | | | | | | | | | | | | | |
| SE Quadrant | Area Source | Area Source | Resuspension | Pu-239 | ⁱ | NA | NA | NA | None | 1 | NA | NA | NA | 0.0E+00 | NA | NA | NA |
| SITE 300 DIFFUSE SOURCES | | | | | | | | | | | | | | | | | |
| Diffuse sources consist of resuspension of depleted uranium from historical explosive tests. The SW-MEI isotopic ratio for S300 in 2012 was 0.0072 and is the ratio for naturally occurring uranium. There were no atmospheric depleted uranium shots in 2012. | | | | | | | | | | | | | | | | | |
| Site 300 | All | Area Source | Soil resuspension | U-238 U-235 U-234 | ^j ^j ^j | NA | NA | NA | None | 1 | NA NA NA | NA | NA | 0.0E+00 | NA | NA | NA |
| NOTE: To convert curies to becquerels use 1 Ci=3.7E+10 Bq and to convert millirem to sieverts use 1 Sv=1.0E+05 mrem. | | | | | | | | | | | | | | | | | |
| ^a Gross alpha and Gross beta emissions are continuously monitored at the stack. | | | | | | | | | | | | | | | | | |
| ^b Because monitoring takes place after HEPA filtration, an unabated EDE cannot be determined from the monitoring data (see discussion in Section 2, Emissions Data). | | | | | | | | | | | | | | | | | |
| ^c Stack emissions have been combined as permitted by the EPA/DOE Memorandum of Understanding. | | | | | | | | | | | | | | | | | |
| ^d Tritium HT and HTO emissions from the stack are continuously monitored. | | | | | | | | | | | | | | | | | |
| ^e Air emissions are continuously sampled at the post-HEPA-filter atmospheric discharge points, although potential emissions are low enough that stack monitoring is not required per the NESHAPs 40 CFR 61 regulations. | | | | | | | | | | | | | | | | | |
| ^f Gamma Emissions are continuously monitored at the stack | | | | | | | | | | | | | | | | | |
| ^g Radioiodines are continuously monitored at the stack | | | | | | | | | | | | | | | | | |
| ^h The unabated EDE shown is only for the tritium source term. | | | | | | | | | | | | | | | | | |
| ⁱ Continuous monitoring for radioactive particulate is in place at surveillance locations | | | | | | | | | | | | | | | | | |
| ^j Continuous monitoring for tritium is in place at surveillance locations | | | | | | | | | | | | | | | | | |

A.2.2 SITE 300 TOWER

5.62702

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0.04210.04310.02880.01360.01250.01370.02200.04010.07710.10590.07750.06320.06930.16680.18030.0441
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5.6095.2366.0053.6383.0332.3571.9652.5775.5986.6695.4454.9776.3498.5179.7415.511
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0.0000 0.0057 0.0392 0.8692 0.0562 0.0297 0.0000
0.0000 0.0078 0.0439 0.5814 0.2661 0.1008 0.0000

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ATTACHMENT 3: POPULATION DATA

The source of the geographic population distribution data used for this report is Oak Ridge National Laboratory (ORNL) LandScan™ 2010 data and ESRI ARCMAP software. The data are placed into an annular grid that is created from sixteen 22.5-degree sectors centered on the cardinal wind directions and five distances spaced at 16 km to a total 80-km radius. In deriving the population for each site, the ORNL data set is input into ESRI ARCMAP with the 80-km grid for the Livermore site centered at 37.686 N latitude, -121.7045 W longitude (near the center of the site) and Site 300 centered at the 52-m meteorological tower located at 37.675 N latitude, -121.541 W longitude. The first line of the input file is informational. Distances are shown in the second row. Population data begin in the third row starting with direction, N. There are 20 spaces reserved for each direction no matter how many are used; i.e., the next direction, NNW, starts approximately half-way through the fifth row, 21 values after the first value.

